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Distribution of Potentially Toxic Soil Elements along a Transect across Kazakhstan

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Abstract

The present study aims to investigate the distribution of selected potentially toxic elements (PTEs) in Kazakhstan's topsoils. Soil samples collected across a north-south gradient (n=40) near main highways connecting major residential/industrial areas were characterized for their As, Cd, Co, Cr, Cu, Mn, Ni, Pb, and Zn content as well as for soil physio-chemical properties. The majority of the soils had neutral pH (no significant relationship between pH and PTE concentrations). The soil organic carbon was higher at the northern and farther southern parts of the transect (along with higher concentrations of PTEs in soils). As, Mn, and Ni concentrations in soils were elevated in comparison to relevant background concentrations. Critical concentrations of As, Cd, Co, Mn, and Ni (with respect to regulatory limits) were found at multiple locations, with As being particularly of potential concern (range: 8.7-42 $mg \times kg^{-1}$). The distance from the nearest settlement seems to influence the soil PTE concentrations, however the relationship is not statistically significant. In total, eight locations had statistically outlier PTE concentrations for Cd, Mn, Ni, and Zn. The overall results were comparable to similar studies across the world except that the Pb content of the study soils was less elevated. Studies on site characterization and human health risk assessment covering identified hotspots and PTEs are recommended.

Key words: arsenic, heavy metals, Kazakhstan environment, site characterization, soil pollution

1. Introduction

The Republic of Kazakhstan is situated in Central Asia and is the ninth largest country in the world in terms of its land area, covering more than 2.7 million km^2. Kazakhstan has vast quantities of land resources (mainly minerals, oil, and gas). As a result, since gaining its independency, its government has mainly depended on the extraction of oil, gas, and minerals as well as the operation of related industries for the country's economic growth. As a result, Kazakhstan is the political leader among Central Asian countries with an economy showing the strongest performance with a continuous growth momentum (Makhmutova, 2018).

The current economic growth in Kazakhstan is mainly fueled by the exploitation of its land resources and accompanying booming sectors of construction and government services, and this may have negative effects on the environment such as problems related to contamination including of water and soil (Kuroda *et al.* 2005). Kazakhstan's soils not only show a tendency of being affected from land degradation, desertification, and salinization due to natural and anthropogenic factors but also may be under environmental strain due to contamination by heavy metals. This could negatively affect public health as well as economy via a reduction in land productivity, stability, functions, and services depending on the exploitation of its natural systems.

A limited number of localized studies have investigated environmental problems in different regions of the country, some also focusing on the potentially toxic elements (PTEs) in soils: Mueller *et al.* (2014) investigated the level of heavy metals in oil-polluted soils in Atyrau region, the reported concentrations of measured Pb, Co, Ni, Cd, Cu, Zn, V, and Mo were 2-12 times higher than the maximum permissible concentrations (MPCs) stated in legislation. The researchers state that the oil-polluted soil became an accumulator and a storage for hazardous forms of the heavy metals which could migrate to the Caspian Sea and thus could cause problems of larger scale. Also, they investigated hazardous mobile forms of Cu, Pb, Ni, Zn, and Cd in large rice irrigation farms in southern Kazakhstan, which showed that heavy metal concentrations impacted soil fertility in agricultural areas where mobile forms of measured heavy metals accumulated in soil profile, making hazardous metals potentially available to crop plants and in drinking water. Ramazanova (2016) investigated Pb, Zn, Cd, Cu, and Fe in soils near central heating power plants at the different distances (50-5000 m) in Almaty: The levels of Fe and Zn in soils complied with the MPCs; however, the Cd and Pb concentrations were five to ten times higher than the MPCs. Bayandinova *et al.* (2018) investigated heavy metals, hydrocarbons, nitrogen and carbon oxides, and organic compounds in air, water resources, and soil at East Kazakhstan region. Specifically, Cd, Cu, Pb, and Zn were investigated in soils near smelters at distances from <1 km to 3 km near Ust-Kamenogorsk, Semey, and Ridder. Elemental concentrations were higher than the MPCs in all investigated districts of Ust-Kamenogorsk in spring and autumn campaigns, the same was also observed in Ridder and Semey. Specifically, the concentrations of Pb, Cd, and Zn were noted as very high one km near the large power plants. Muzychenko *et al.* (2017) studied the Pb pollution in the roads of Almaty and found high concentrations attributed to traffic as well as the industrial and geological history of the region. Finally, Mukasheva *et al.* (2013a, 2013b) monitored Cr, Cu, Cd, Pb, Zn, Ni, V, and Mn pollution near Temirtau, Balkhash, and Karaganda towns. All these cities are industrial centers with large smelters and plants near or inside the city boundaries such as Karaganda Steel Mill and chemical plants "Jambul Cement" and "ArcelorMittal" in Temirtau. Their comparative data analysis showed that the concentration of heavy metals in all soil samples exceed the MPCs and heavy metals could accumulate in soil and crop plants.

Although a number of localized studies already indicate environmental adverse impact from anthropogenic activities, a systematic study that is reporting soil quality conducted on a larger scale is missing in the literature. Although Kazakhstan is the economical leader of Central Asia and one of the major developing countries around the world, the environmental consequences of this expansion has not yet been well studied. As a part of a large study that aims to investigate Kazakhstan's soils in terms of its physical and chemical properties (Yapiyev *et al.* 2018), contamination status, and nutrient potential, the present study aims to investigate the distribution of selected PTEs (As, Cd, Co, Cr, Cu, Mn, Ni, Pb, and Zn) in topsoils of Kazakhstan collected through a north-south transect (n=40, sampling sites approx. 50 km apart) near highways connecting numerous major residential/industrial landmarks of the country. We aim to identify hotspots as well as relationships between individual PTE concentrations and site/soil properties. Establishing a baseline about the current status is important as this not only help identify potential problem areas but also could be used to compare with the future monitoring and later to predict the dynamics of the change.

2. Materials & Methods

2.1. Sampling Area, Sampling and Sample Handling

Soil samples (n = 40) have been collected in September 2016 (Fig. 1) from sampling sites located a minimum of 50 m away from the nearest road. The sample from the northernmost point was collected near Petropavlovsk city (5454'N, 6906'E) and the southernmost sample was collected near Chu river (4255'28''N, 7311'6''E). During sampling, following the removal of the litter layer if present (approximately the top 2 cm), drilled samples were cored from approximately 0-15 cm. The samples, then, were air-dried in the laboratory, sieved to < 2 mm, and stored at room temperature until analyzed (see also Methods section in Yapiyev *et al*. (2018)).

2.2. Laboratory Analyses

Soil pH was measured in soil suspension using 8107UWMD Ross Ultra pH/ATC triode electrode (Thermo Fisher Scientific, USA) and an Orion 013010MD conductivity cell electrode (Thermo Fisher Scientific, USA). Soil organic carbon (SOC) was measured using a C/N dry combustion elemental analyzer (Multi N/C 3100, Analytic Jena). Samples were pre-treated on the combustion boat by adding 100 μL of H_3PO_4 (30-40% w/w) to 100 mg milled soil to dissolve carbonates first. The samples were then dried overnight at 70 °C and finally subjected to combustion at 950 °C under 14 $L \times min^{-1}$oxygen flow (see also Methods section in Yapiyev *et al.* (2018)).

The selected PTEs were measured using ICP-MS iCAP RQ. Sample digestion was first conducted via a microwave system (Multiwave PRO by Anton Paar). Soil samples were dried for 48 h, crushed, and sifted through 150 μm sieve. Then, 1 g of soil was weighed accurately at 0.1 mg (Radwag XA52/Y) and then was transferred into a vessel. 9 mL HCl (37% w/w) and 3 mL of HNO_3 (68-70% w/w) were added separately to the vessel, and samples were digested at 140 °C for 40 min. After the digestion has been complete, all vessels were cooled, then the digestates were filtered through Whatman No. 1 or equivalent filter paper (11 μm pore size). After cooling and filtering, solutions were collected in a volumetric flask and then diluted to 50 mL. The reagent blanks were carried out in parallel with each experimental analysis, All analyses were conducted in duplicates. Furthermore, for quality control purposes the reference material BGS 102 was digested and the concentrations of PTEs were measured using ICP-MS iCAP RQ. The concentrations of measured Cr, Cu, Ni Pb, Zn were close to certified values (i.e. within 20%), except for Cd where the difference between measured and certificated (0.275 ± 0.182 mg $\times$ kg^{-1}) concentration was about 40%.

2.3. Data Processing and Statistical Analyses

Apart from calculations performed for determining selected descriptive statistics (average, minimum, maximum, standard deviation, coefficient of variation, skewness, kurtosis), The Pearson correlation tests have been conducted to check the relationship between (a) individual PTE concentrations, (b) PTE concentrations and soil physio-chemical properties, and (c) PTE concentrations and distance of sampling sites from nearest settlement (a settlement defined as a residential zone with population > 1,000). Sampling points were also categorized into two groups according to their distance from the nearest settlement; the first including points relatively close to the nearest settlement (distance < 10 km, n = 20) and the second consisting of points relatively far from the nearest settlement (distance > 10 km, n = 20) where exact distances are presented in the Supplementary material (Supplementary Table 1). These groups were compared via two sample *t*-test in order to see whether there are differences between PTE concentrations. Shapiro-Wilk test along with Q-Q plots have been employed to check the distribution of data with the specific purpose of identifying outlier PTE concentrations. Furthermore, a box plot visually representing the data for potentially toxic elements has been prepared (Fig. 3) via software (IBM SPSS 25). Outliers have also been determined. which consist of points with concentrations of PTEs larger than 1.5 × IQR (interquartile range, the difference between the third and the first quartiles).

3. Results and Discussion

3.1. Soil Properties

The pH of the samples (Supplementary Table 1) ranged from 5.32 to 9.18. The majority of the samples had neutral pH (between 6 and 8) except two samples that can be categorized as acidic (with pH < 6, sampling locations: S7, S10) and 11 samples that could be classified as alkaline (with pH > 8: S25-S32, S35, S37-S38). The pH has a significant effect on the mobility of trace metals in soil such that at a lower pH the sorption capacity of soil is lower than that at neutral or alkaline pH, Consequently, the mobility of most part of metals decreases from acids soils to alkaline. Soil samples from the present research are mostly neutral therefore the expected mobility of metal forms is generally at the lower end. There was also a trend that through the north-south transect (Fig. 2, based on data from Yapiyev *et al.* (2018)), the soils samples become more alkaline from north to south which could be linked to more arid climate conditions and dominance of calcareous soils (Slessarev *et al.* 2016) .

The amount of SOC in soil (Supplementary Table 1) influences, among many other things, the mobility and bioavailability of various elements by binding them into its complex structure and thus limiting their mobility at higher SOC values. The SOC content of study soils ranged of 0.38% to 4.09% showing that the study samples collected through the transect were mainly mineral (inorganic) soils. Through the north-south transect, higher SOC values (> 2%) identifying potentially organic soils have been observed specifically in the north (S3, S5-S9, S12, S14, S16) and farther south (S34, S36-S40). These zones are also inside the country's two important arable regions, therefore, such an observed specific elevated profile of SOC would be expected.

3.2. Concentrations of PTEs in Soils

The levels of potentially toxic elements (PTEs) were first compared with the background concentrations of Canadian and Chinese soils (Tables 1 and 2). The background levels may be useful in order to estimate the differences between natural content of metals including their geochemical composition as source and the fraction coming from contamination sources (Santos-Frances *et al.* 2017). The background concentrations of Canadian (MDDELCC 2019) and Chinese (Huamain *et al.* 1999) soils were used since in certain parts of these countries, the climate as well as the conditions that the surface soils had been developed were comparable to those in Kazakhstan.

The comparison with the Canadian soil background levels showed that particularly As concentrations were above background levels such that all of the samples from the present study had As concentrations larger than 6 $mg.kg^{-1}$, corresponding to an average As concentration in the study soils was 20.8 $mg.kg^{-1}$. Considering the other PTEs, Mn and Ni average concentrations were also higher than corresponding background values (34 of 40 points for Mn and 29 of 40 points for Ni exceeded the background value). The levels of Co, Cr, Cu, and Zn were comparable to the Canadian background levels (i.e. with comparable average vales) whereas Pb concentrations were lower (only five points exceeding the background level) and Cd concentrations were much lower (all points below the background level) than the Canadian background concentrations.

The comparison of PTE concentrations from the present study with Chinese soil background values (which reported values are lower than the Canadian background values except for As) showed that the found concentrations were higher for As, Cd, Cr, Cu, Ni, and Zn; and were comparable for Pb. No element had systematically lower concentrations than the background values (no background values were present for Co or Mn therefore no comparison could be made).

Overall, the comparison of the PTE concentrations in the study soils with the background concentrations of soils of China and Canada indicated high concentrations of especially As and of Mn and Ni. The evidence regarding the elevated Cd, Cr, Cu, and Zn concentrations was less clearly cut.

The results from the soils of the present study were also compared with regulatory standards of Kazakhstan and Russia (which share the same regulatory limits), China, and Canada for soils. Regarding the regulatory standards of Kazakhstan (MEPRK 2004) and Russia (RPR 2006), for As, Co, Cr, Ni, and Zn, the measured concentrations exceeded the maximum permissible concentrations (MPCs) in every single sample. The majority of the samples also exceeded the criteria for Cu (35 of 40 samples) and Pb (22 of 40 samples) whereas for Mn and Cd, less samples had concentrations higher than the MPC (19 and 9, respectively, of 40 samples). That being said, many MPCs presented in Kazakh and Russian regulations seem low or very low even in comparison to natural background concentrations of these elements as in the cases of: As (MPC: $2\mathrm{mg} \times \mathrm{kg}^{-1}$), Cd ($0.5\mathrm{mg} \times \mathrm{kg}^{-1}$), Co ($5\mathrm{mg} \times \mathrm{kg}^{-1}$), Cr ($6\mathrm{mg} \times \mathrm{kg}^{-1}$), Cu ($33\mathrm{mg} \times \mathrm{kg}^{-1}$), Ni ($4\mathrm{mg} \times \mathrm{kg}^{-1}$), and Zn ($23\mathrm{mg} \times \mathrm{kg}^{-1}$). Since we could not find any supporting documents and references that provide a scientific rational for these regulations it is not possible to evaluate the scientific soundness of these limits. Consequently, it may be recommended to not to depend on these comparisons alone for evaluating the levels of PTEs of the study soils.

When the results from the present study have been compared to the regulatory standards in Canada for residential soils (MDDELCC 2019), the only element with systematically high concentrations in topsoils was Mn (concentrations exceeding the limit at 34 of 40 points). Also, As, Co, and Ni concentrations were high in five, four, and six points, respectively. This comparison identified four critical sampling locations with elevated PTE concentrations indicating possible contamination with three or more elements: S8 (with As, Co, Mn, and Ni), S35 (with Co, Mn, and Ni), S36 (with As, Co, Mn, and Ni), and S39 (with As, Mn, and Ni).

A similar comparison with the Chinese environmental quality standards (MEPPRC 1995) showed that Cd and Ni concentrations were higher than the stated norms at the majority of the points (24 and 29 points, respectively, out of 40). This is mainly because the Chinese regulation has stricter limits for Ni and Cd in comparison to the Canadian regulation. Parallel to the above comparison, the As concentrations were higher than the norm at five locations. This comparison identified seven sampling locations with elevated PTE concentrations indicating possible contamination with three or more elements: S7 (with Cd, Cr, and Ni), S8 (with As, Cd, and Ni), S9 (with As, Cd, and Ni), S19 (with As, Cd, Ni, and Zn), S28 (with Cd, Cu, and Ni), S36 (with As, Cd, and Ni), and S39 (with As, Cd, and Ni). The common locations with the comparison to Canadian standards were S8, S36, and S39.

3.3. Analysis of Concentrations and Potential Hotspots

The descriptive statistics for PTEs in the study soils (Table 2) provided no average values or ranges at the extreme ends for the selected PTEs. As discussed previously, a comparison of the elemental averages to Canadian and Chinese environmental norms indicated As and Ni (by both regulations) as well as Cd, Co, and Ni (according to one of the regulations) as PTEs exceeding norms at some locations. The coefficient of variation values fell in a normal range except that it was higher for Pb, possibly due one outlier point with high concentration (S11). The skewness of the data was to the left for all PTEs, indicating a pooling of values at the lower range while a few points with higher concentrations extended the distribution tail to the right end, which could be reasonably expected for the soil PTE concentration data from this site. Finally, the data were leptokurtic for the majority of the elements as expected, with the exception of Cr and Cu having flatter distribution than the other elements which may indicate anthropogenic disturbances.

Inter-element relationships may provide information on the sources and pathways of PTEs. According to the Pearson correlation coefficients (Table 3), the concentrations in eight of the nine selected PTEs (with the exception of Pb) are significantly correlated with each other at $p < 0.01$ level. It is known that the behavior of metals in soils are controlled by a number of key parameters (USEPA 1992) including the soil's pH and cation exchange capacity (which is mainly defined by soil organic matter and clay content of the soil) when cations are of concern. As a result, it may be expected to observe such a correlation between elemental concentrations in a set of soil samples as certain soil physio-chemical properties control the mobility and retention of a variety of PTEs in a similar fashion. The fact that Pb has a set of correlations with other PTEs with lower and/or with less/not significant values may be due to different reasons. In terms of its affinity to soils and soil constituents, Pb has a high relative order of sorption (USEPA 1996) and consequently it is one of the least mobile heavy metals in soils. As a result, other PTEs in soil might have exhibited relatively higher mobilization rates in the past which leading to a reduction in their soil concentrations with time. Another possibility is that Pb might be introduced to the environment from overland traffic activities (the samples locations have been accessible sites from nearby highways) therefore shows a different concentration profile than the other PTEs. This, however, is unlikely as Pb tends to accumulate mostly within the nearby soils, particularly within the first 10 m whereas the sampling locations were located a minimum of 50 m away from the nearest road. That being said, the overland traffic activities introduce to the environment not only Pb but also Cu and Zn (Guney *et al.* 2010), and in the present study Pb is also correlated with these PTEs at 0.01 level, therefore, this may require further investigation in the future.

The correlation between PTEs concentrations and soil pH & SOC (Table 4) showed that although the concentrations of the majority of PTEs showed an inverse correlation with pH, the values were not statistically significant. Therefore, the pH of the soils was not a major parameter controlling the PTE concentrations in soil. On the contrary, the SOC values were positively and significantly correlated with PTE concentrations. The soil organic matter significantly contributes to cationic and anionic exchange capacities, therefore may be expected to increase the retention of PTEs in soils.

For the two categories of points according to their distance from the nearest settlement (population > 1,000), the points that are relatively closer to the nearest settlement (< 10 km, n = 20) had their average concentrations of the majority of the PTEs (Table 5) higher than that of the points that are relatively farther (>10.0 km, n=20). However, the two sample *t*-test indicated that the difference between the concentrations of PTEs was only significant for Cr. Although it first seems that the settlements have an impact on the topsoil concentrations of PTEs, this impact is not statistically significant. An analysis of Pearson correlations between the concentrations of individual PTEs and the distance of the sampling location from the nearest settlement (Table 6) also indicated negative correlations for the majority of the elements: however, the values were low and were significant at 0.05 level only for three elements. Therefore, it is not possible to talk about a significant impact of settlements on the PTE concentrations of topsoils. Such a significant impact may not have been observed due to the fact that some of the settlements are not large enough to have a major impact on nearby soils via anthropogenic activities. It is also possible that the impact of anthropogenic activities may not be significant due to relatively large distances between settlements and sampling locations.

Shapiro-Wilk's normality test has been employed (Table 7) together with Q-Q plots and histograms with the specific purpose of identifying outliers having excess concentrations of PTEs. The data showed normal distribution only for As, Cr, Mn, and Zn. An examination of test results together with Q-Q plots and histograms revealed eight locations with outliers: one location with three outliers (S36 for Cd, Mn, and Ni), two locations with two outliers (S10 for Cd and Mn, S32 for Cd and Zn), and five locations with one outlier concentration (S7, S9, S11, S19, and S28). A more detailed discussion is presented below regarding the identification of specific sampling locations of concern in terms of elevated PTE concentrations. Finally, the box plot analysis identified similar locations with outlier concentrations: at S10 (Pb), S11 (Pb), S32 (Cd), S35 (Co), and S36 (Co, Mn, Ni).

Based on the previously presented comparison with the background concentrations and the regulatory limits from selected countries, the PTEs of concern for the present study could be identified as As, and to a lower extent Cd, Mn, and Ni. In terms of locations, a comparison to regulatory limits and the statistical analyses yielded varying results (Canada: S8, S35, S36, S39; China: S7, S8, S9, S19, S28, S36, S39; statistical analyses: S36, S10, S32, S7, S9, S11, S19, S28). At the end, it is possible to identify five locations as potential hotspots: The location S36 has been unanimously identified as a potential hotspot. Similarly, S8 and S39 are two other potential hotspots identified by the comparison to regulatory limits. Finally, it is worth to consider S10 and S32 as potential hotspots as they are pointed by the statistical analyses having outlier concentrations at each location. It is interesting to note that none of these three points are near large settlements or under the direct nearby influence of industrial activities. The elevated concentrations of some PTEs at these locations might be due to natural sources or might result from long-distance impact of anthropogenic activities such as smelting. It should be noted that the sampling locations have been selected on a random basis to represent the topsoil concentrations of selected PTEs and a direct investigation of contamination sources for PTEs was not the major goal. Therefore, at these five locations (S36, S8, S39, S10, and S32), further investigation and site characterization involving multiple sampling locations and taking potential human exposure into account is recommended.

Among five points identified above as potential hot spots for their elevated PTE concentrations by a comparison to regulatory limits and/or via statistical means, their soil types included chernozem (S8, S10), arenosol (S32), regosol (S36), and umbrisol (S39). Chernozems are soils rich in organic matter commonly found in the steppes of Eurasia up to Siberia. The SOC for S8 and S10 were 2.41% and 1.63%, respectively. The elevated concentrations of PTEs in S8 (As, Co, Cd, Mn, and Ni; according to regulations) and S10 (Cd, Mn, Pb; identified as outliers via statistical analyses) may be attributed to cation retention mechanisms from organic matter limiting the mobility of elements. Umbrisols are also steppe environment soils, characterized by a surface layer that is rich in humus. Similar to S8 and S10, this may explain high concentrations of PTEs in S39 (As, Cd, Mn, and Ni; according to regulations). Regosols are weakly developed soils, a characteristic of eroding landscape, and it not easy to comment on their properties due to unconsolidated material they are formed and a lack of soil horizons. However, regarding the presence of PTEs, S36 is close to other sampling locations with umbrisol soil type (S37 to S39), therefore, high concentrations of PTEs in S36 (As, Co, Cd, Mn, and Ni; according to regulations) would be partly explained by the similarities to the concentrations in neighboring sampling points (e.g. S39). Arenosols, on the contrary, may be expected to have generally low PTEs due to their sandy nature accompanied by low humus and clay content. Therefore, outlier Cd and Zn concentrations at S32 warrant further investigation where possible explanations include contaminated dust fallout from industrial facilities or of geogenic origin e.g. relatively common sulfide minerals greenockite (CdS) and wurtzite (ZnS).

3.4. Comparison to Studies Investigating Similar Sites

Anthropogenic sources affect the PTE concentrations in soils. A study on the general patterns of element abundances in urban soils pointed out that the overall pattern of elements in Earth's soils tends to mainly repeat those in the Earth's crust (Alekseenko *et al.* 2014). It also indicated that certain elements tend to be more abundant in urban soils than the Earth's soils in general, including some of the PTEs (As, Cd, Co, Cu, Pb, and Zn) subjected in the present work.

A comparison of the results from the present study to studies in China (where rural areas with similar climate to the present study sampling locations and under influence of nearby anthropogenic activities are common) indicated similar findings. Cheng (2003) reviewed the contamination by heavy metals (focus on Cd, Cu, Pb, and Zn) in different areas of China including rural and agricultural areas. The background levels of heavy metals in soils were low, but human activities have been shown to pollute soil, water, and air where metals may be transferred to plants and food; three identified main sources of contamination being industrial emissions, wastewater, and solid waste. The reported heavy metal concentrations were within the range in the present study, with the exception of Cd being higher and Zn and Cr being also slightly more elevated. Another review of heavy metal contaminations in urban soils, road dusts, and agricultural soils from China (Wei *et al.* 2010) showed that among selected elements (Cr, Ni, Cu, Pb, Zn, As, Hg, and Cd), the geoaccumulation index of urban soils showed Cu, Pb, Zn and Cd enrichment and of agricultural soils had elevated concentrations of Cd, Hg, and Pb due to anthropogenic activities with various sources (mining, sewage sludge, pesticides, fertilizers, traffic emission, electroplating plant, spring factory, band steel factory, leather factory, petrochemical complex, etc.) noted as the main sources of trace metals contamination. The contents of PTEs were in general comparable to the present research. Liao *et al.* (2007) has performed a geochemical survey of soils of Jiangsu Province, China for 54 elements in 103,000 top-soil samples and presented similar results regarding PTE concentrations. They reported that for eight PTEs (As, Cd, Hg, Pb, Ni, Cr, Cu, and Zn), the measured concentrations of these PTEs were higher than their natural background at 40% of the samples (with over 10% of the land being classified as contaminated). Finally, Zhao *et al.* (2010) considered anthropogenic activities as the reason of PTE contamination of Yangtze River Delta region. They analyzed selected PTEs (Cu, Zn, Pb, Cr, Ni, Cd, and Hg); among these, the concentrations of Cu, Zn, and Pb significantly exceeded background levels.

Other studies of comparable nature to the present study have reported similar results to the present study with a few caveats. The levels of some PTEs (As, Cd, Co, Cu, Fe, Ni, Pb, Mn, Mg, and Zn) in soil near smelters in the Sudbury region in Ontario, Canada (a region with comparable climate and soils to the present study) were monitored (Nkongolo *et al.* 2008). The concentrations of PTEs were within the limits set by Ontario Ministry of Environment and Energy (OMEE) guidelines (which are similar to MDDELCC guidelines from Quebec, Canada used in the present study), even in sites within the vicinity of the Falconbridge Smelters. A comparison to the concentrations in the present study showed similar PTE levels except for higher Co, Mn, Ni, and Zn content in the present research, where presumably a strict enforcement of guidelines in Ontario may have led to such a difference. The concentrations of PTEs reported by Nezat *et al.* (2017) in urban soils of Spokane, Washington also reported similar results to the present study with the exception that they reported elevated Pb concentrations which has been likely due to lead-based paint or vehicle emissions in urban zones. A study conducted on urban soils of Yerevan city, Armenia (Tepanosyan *et al.* 2017) reported similar concentrations of PTEs and some enrichment in urban zones with the exception of clearly elevated Pb concentrations attributed to the historical pollution in the study zone. These findings on Pb are also parallel to the findings of Turner *et al.* (2018) in urban soils of Plymouth, UK who reported elevated Pb due to contamination by paint. Also, Pb contamination tends to decrease to the background value at about 70 m or more roadside distance (Yan *et al.* 2013). This is in agreement with our findings as the soils of the present study did not exhibit an extensive contamination by Pb because all samples were taken from locations with relatively limited impact of transportation (sampling locations at least 50 m away from the roads) or urban activities (many sampling locations distant from urban settlements). Still, such an impact might be important for other PTEs as different studies indicate that both local and distant sources of anthropogenic emissions may still lead to enrichment of PTEs in non-urban soils e.g. in national park soils in Poland by Pb, Zn, Cu and Mn (Mazurek *et al.* 2017) and in soils from forests and protected areas in Romania by Pb, Zn, Cu, Cr, and Cd (Ungureanu *et al.* 2017). Finally, a very large soil survey covering the entire Europe and analyzing more than 15,000 surface soil samples have proposed 28.3% of the total surface area of the EU for further assessment because one or more of the elements (among As, Cd, Cr, Cu, Hg, Pb, Zn, Sb, Co and Ni) exceed the applied threshold concentration, identifying both natural background and historical/recent industrial and mining areas as potential sources. Similarly, the present study identified eight locations with at least outlier elemental concentration as pointed out by the statistical analyses (corresponding to 20% of 40 sampling locations).

4. Conclusions

As a rapidly developing country, Kazakhstan relies on its vast land resources of minerals, oil, and gas together with related industries, which may create a major burden on its environment. Soil is an important and now scarce resource and its contamination by potentially toxic elements (PTEs) may limit its usability for agricultural and urban purposes. The present study investigated the distribution of selected PTEs (As, Cd, Co, Cr, Cu, Mn, Ni, Pb, and Zn) in topsoils of Kazakhstan collected through a north-south transect at 40 locations. The analyses have shown that the soils from the study area were mostly neutral in terms of pH, with a tendency of having more alkaline samples as moved from north to south. The statistical analyses didn't indicate a significant relationship between pH and the PTE concentrations. The soil organic carbon was higher at the northern and farther southern parts of the sampling transect and these locations also had higher concentrations of PTEs in soils. A comparison of PTEs to background concentrations of soils in two countries with similar climate and soil properties (China and Canada) indicated elevated concentrations of As, Mn, and Ni in soils. A comparison to regulatory limits of China, and Canada also indicated critical concentrations of As, Cd, Co, Mn, and Ni in multiple locations (Some of the regulatory limits of Kazakhstan and of Russia were very low and the scientific basis for establishing these limits was not disclosed; thus, conclusions based on these limits are not emphasized here). Although the distance of the sampling locations from the nearest settlement (population > 1,000) seemed to influence the PTE concentrations, the relationship was not statistically significant. Further statistical analyses identified eight locations with outlier PTE concentrations: S36 for Cd, Mn, and Ni, S10 for Cd and Mn, S32 for Cd and Zn), and five locations for one element (S7, S9, S11, S19, and S28). Overall, the results of the present study were comparable to previously conducted studies around the world at locations with similar properties with the specific remark that the Pb content of soils was less elevated than some other study locations with a history of anthropogenic impact. Studies aiming the site characterization and human health risk assessment on identified hotspots (particularly at S36 as identified via comparison to legislations and by statistical investigations) and PTEs (particularly for As that is elevated at most locations) are recommended.

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Conflict of Interest:

The authors declare that they have no conflict of interest.

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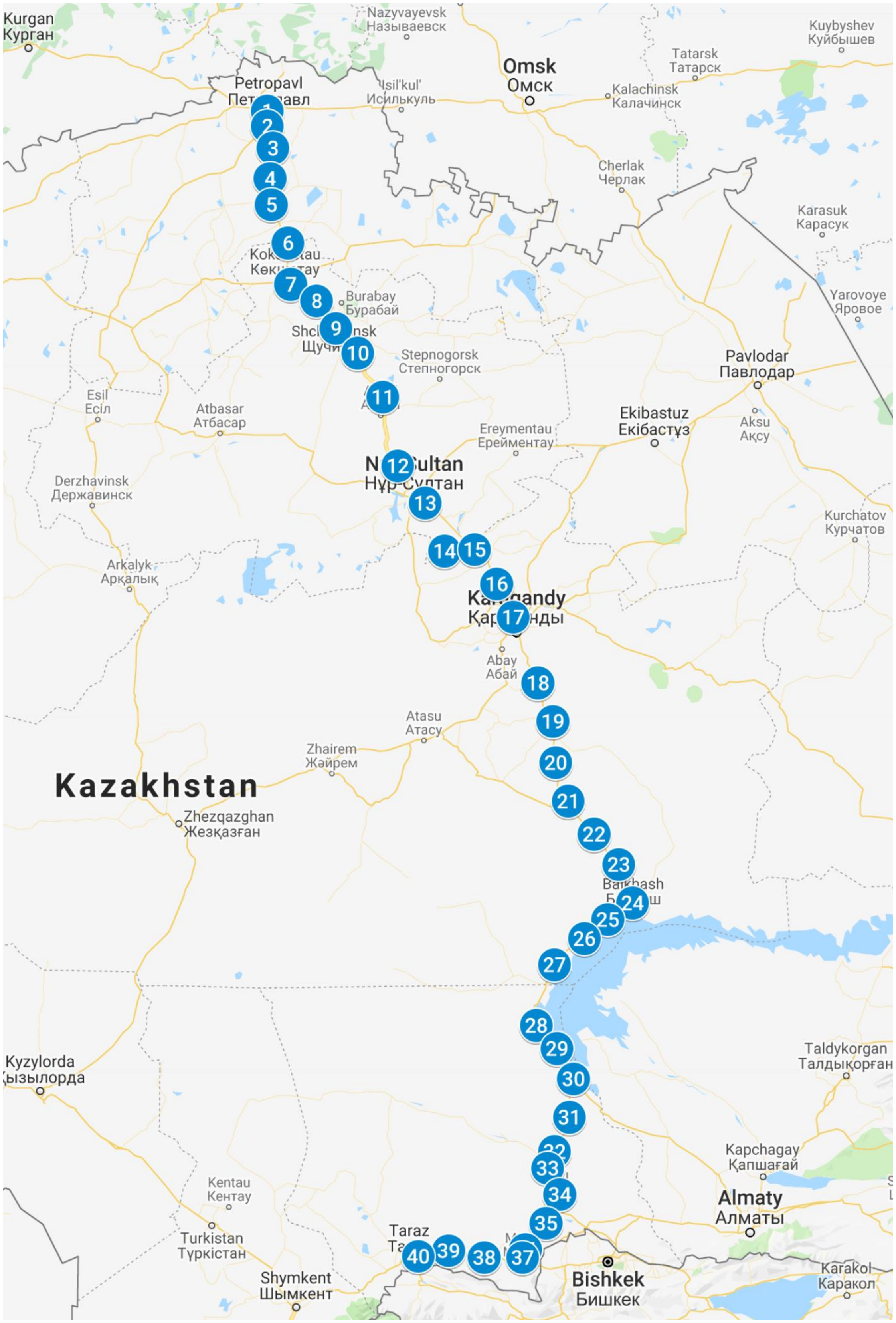

Fig. 1. Study area with locations of sampling sites

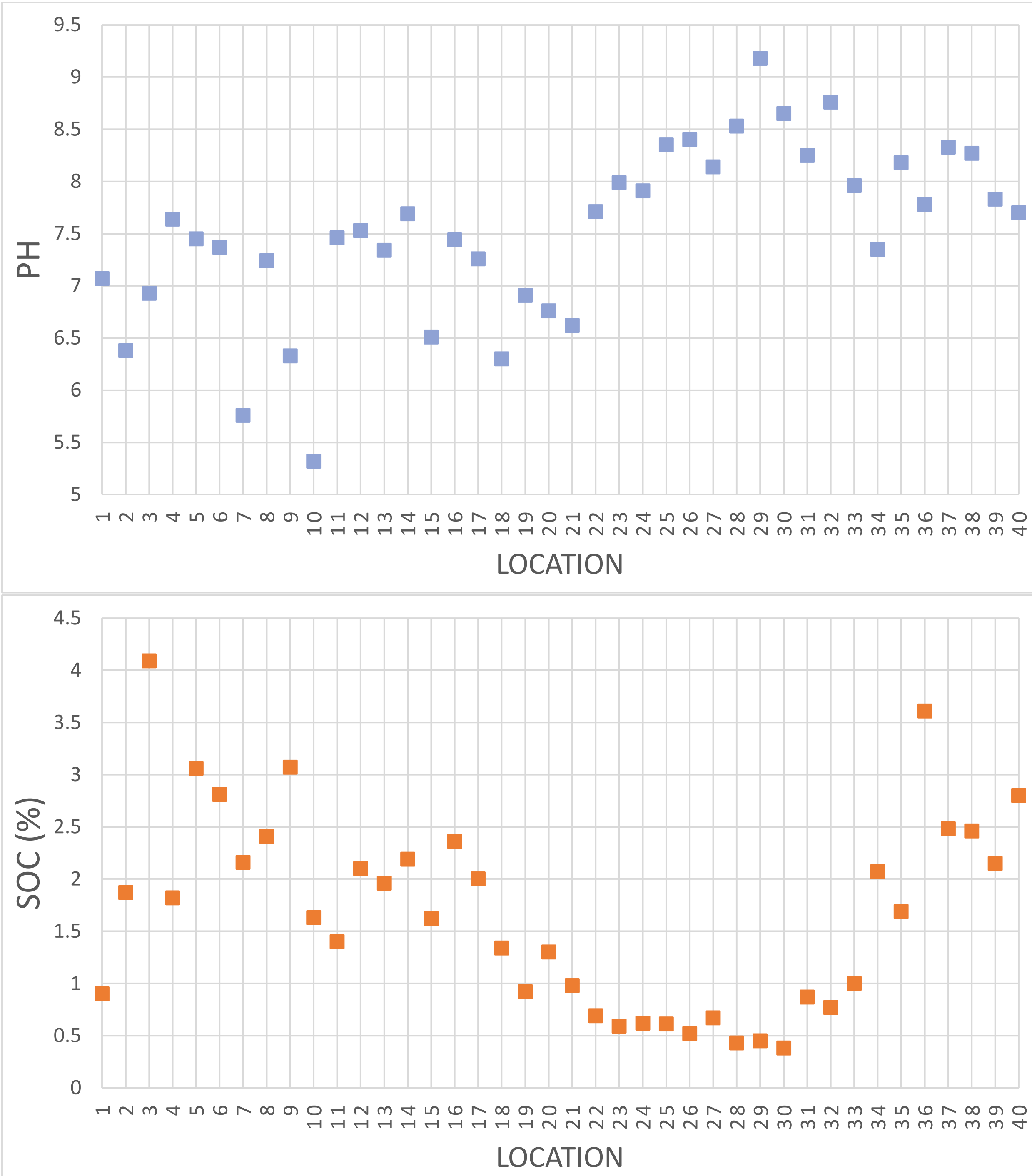

Fig. 2. Soil pH and SOC values (0-15 cm depth) through the north-south transect (based on data from Yapiyev *et al*. (2018))

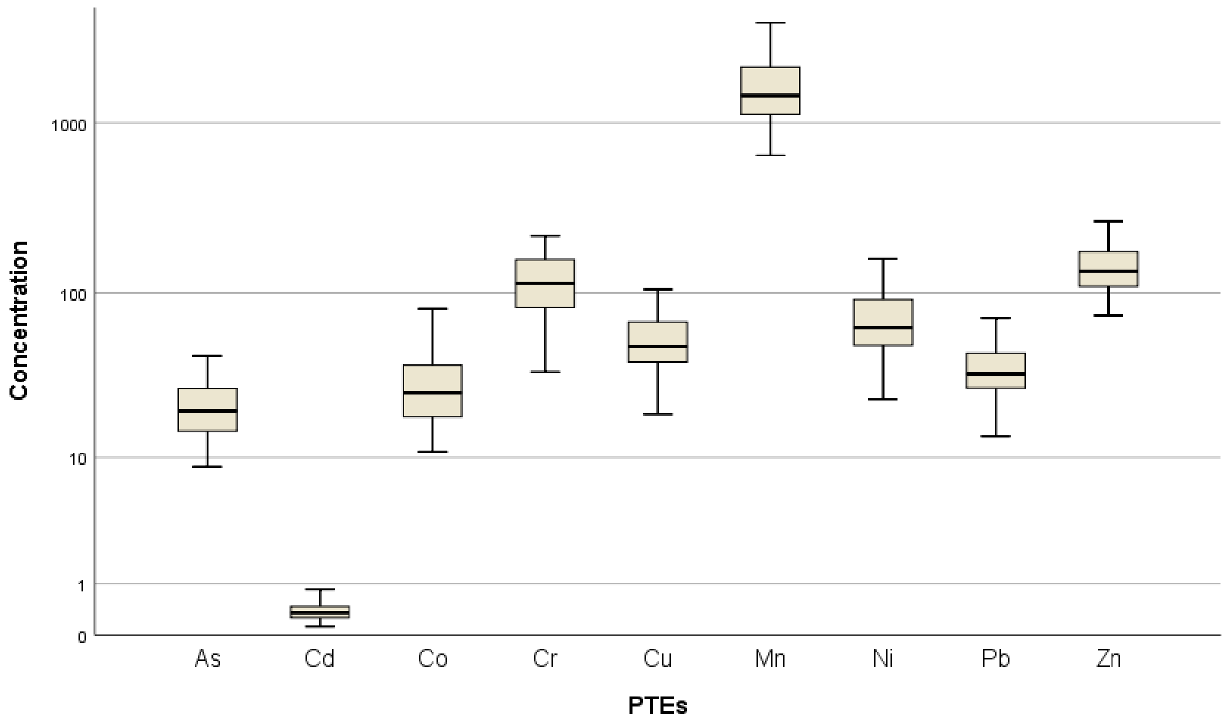

Fig. 3. Box plot for concentrations of PTEs in soil samples (0-15 cm depth) (boxes: first, second, and third quartiles; whiskers: ±1.5 × interquartile range)

Table 1. Range of concentrations of potentially toxic elements (PTEs) in the soils of present study (0-15 cm depth), background levels of PTEs in Canadian and Chinese soils, and regulatory standards for PTEs for Kazakhstan, Russia, Canada, and China (mg.kg^{-1})

PTE	**Range for the present study**	**Natural background values (Quebec, Canada) (MDDELCC 2019)**	**Natural background values (China) (Huamain *et al.* 1999)**	**National standards for Kazakhstan (MEPRK 2004) and Russia (RPR 2006)**	**Residential limits (Quebec, Canada) (MDDELCC 2019)**	**Chinese Environmental Quality Standards (MEPPRC 1995)**
As	8.70 - 42.0	6	≤15	2	30	30
Cd	0.12 - 0.85	1.5	≤0.20	0.5	5	0.3
Co	10.8 - 80.6	25	N/A	5	50	N/A
Cr	33.7 - 217	100	≤90	6	250	200
Cu	18.6 - 105	50	≤35	33	100	100
Mn	353 - 3.84E3	1,000	N/A	1,500	1,000	N/A
Ni	23.0 - 159	50	≤40	4	100	50
Pb	8.60 - 161	50	≤35	32	500	300
Zn	23.6 - 264	140	≤100	23	500	250

N/A – not available

Table 2. Selected descriptive statistics for PTE concentrations

PTE	Average	Min	Max	Standard deviation	Coefficient of variation	Skewness	Kurtosis
As	20.8	8.70	42.0	7.67	36.9%	0.59	0.007
Cd	0.38	0.12	0.85	0.18	47.9%	0.88	0.529
Co	29.8	10.8	80.6	15.8	53.1%	1.58	2.753
Cr	120	33.7	217	46.9	39.1%	0.28	-0.762
Cu	53.6	18.6	105	20.8	38.7%	0.65	-0.291
Mn	1.68E3	353	3.84E3	762	45.4%	0.83	0.543
Ni	69.6	23.0	159	28.7	41.2%	0.87	0.875
Pb	38.7	8.60	161	23.8	61.6%	3.62	17.883
Zn	140	23.6	264	46.4	33.1%	0.37	0.910

Table 3. Pearson correlation matrix between individual PTEs

	As	**Cd**	**Co**	**Cr**	**Cu**	**Mn**	**Ni**	**Pb**	**Zn**
As	1	0.481**	0.580**	0.809**	0.699**	0.602**	0.763**	0.307	0.642**
Cd		1	0.449**	0.622**	0.610**	0.623**	0.482**	0.295	0.583**
Co			1	0.699**	0.577**	0.767**	0.859**	0.501**	0.513**
Cr				1	0.594**	0.713**	0.873**	0.321*	0.671**
Cu					1	0.628**	0.623**	0.417**	0.763**
Mn						1	0.745**	0.670**	0.725**
Ni							1	0.293	0.658**
Pb								1	0.424**
Zn									1

* Significant at the 0.05 level

** Significant at the 0.01 level

Table 4. Pearson correlations between PTEs and soil physiochemical characteristics

	As	**Cd**	**Co**	**Cr**	**Cu**	**Mn**	**Ni**	**Pb**	**Zn**	**pH**	**SOC**
pH	-0.213	0.012	-0.070	-0.302	0.086	-0.139	-0.127	-0.046	-0.107	1	-0.350*
SOC	0.516**	0.170	0.549**	0.564**	0.273	0.510**	0.669**	0.128	0.394*	-0.350*	1

* Significant at the 0.05 level
** Significant at the 0.01 level

Table 5. Results for two sample *t*-test between two sets of points with distance < 10 km and > 10 km to the nearest settlement

Element	**Average concentration for d < 10 km ($mg \times kg^{-1}$)**	**Average concentration for d > 10 km ($mg \times kg^{-1}$)**	***p*-value**	***t*-statistic**	**Null hypothesis**
As	22.5	19.1	0.161	1.432	Accepted
Cd	0.41	0.35	0.314	1.021	Accepted
Co	33.3	26.2	0.158	1.439	Accepted
Cr	135	105	0.044	2.087	Rejected
Cu	53.2	54.1	0.893	-0.134	Accepted
Mn	1.80E+03	1.56E+03	0.334	0.978	Accepted
Ni	76.7	62.5	0.118	1.601	Accepted
Pb	36.6	40.8	0.583	-0.554	Accepted
Zn	141	139	0.889	0.14	Accepted

Table 6. Pearson correlations between PTEs and distance of sampling sites to nearest settlement

	As	**Cd**	**Co**	**Cr**	**Cu**	**Mn**	**Ni**	**Pb**	**Zn**
Distance	-0.228	-0.257	-0.327	-0.415	0.060	-0.267	-0.364	-0.051	-0.196
r	0.052	0.066	0.107	0.172	0.004**	0.071	0.133	0.003**	0.038*

* Significant at the 0.05 level

** Significant at the 0.01 level

Table 7. Results for Shapiro-Wilk test and outlier points with PTE concentrations

PTE	*p*-value	W	Distribution [a]	Outlier points [b] (concentration in $mg \times kg^{-1}$)
As	0.150	0.9585	Normal	S9 (42.0)
Cd	0.011	0.9251	Non-normal	S32 (0.85), S10 (0.78), S36 (0.76)
Co	0.000	0.8527	Non-normal	S36 (80.6), S35 (75.5)
Cr	0.246	0.9649	Normal	S7 (217)
Cu	0.050	0.9447	Non-normal	S28 (105)
Mn	0.070	0.9490	Normal	S36 (3.84E+03), S10 (3.30E+03)
Ni	0.046	0.9437	Non-normal	S36 (159)
Pb	0.000	0.6824	Non-normal	S11 (161)
Zn	0.431	0.9725	Normal	S19 (264), S32 (243)

[a] Normality is rejected if $p < 0.050$

[b] determined via Q-Q plots and histograms

Supplementary Table 1. Sampling points with data for nearby settlements, concentrations of selected PTEs (0-15 cm depth), and soil properties from Yapiyev *et al.* (2018

Sample ID	Soil type	Location	Nearest settlement	Distance (km)	Population	Elemental concentration ($mg \times kg^{-1}$)									pH (-)	SOC (%)
						As	Cd	Co	Cr	Cu	Mn	Ni	Pb	Zn		
S1	Chernozem	Road street Potanina	Petropavlovsk city	0	201,446	9.66	0.14	23.3	54.7	18.6	642	30.9	14.0	73.2	7.07	0.90
S2	Chernozem	Highway A 1	Beiterek village	2.7	9,679	13.1	0.14	22.7	67.3	28.8	1.38E3	46.4	26.7	87.9	6.38	1.87
S3	Chernozem	Highway A 1	Astrakhanka village	3.2	6,313	21.2	0.41	37.6	157	64.5	2.32E3	94.4	38.2	186	6.93	4.09
S4	Chernozem	Highway A 1	Amangel’dy village	20.3	7,569	17.8	0.31	26.0	128	39.9	1.21E3	72.0	27.7	99.5	7.64	1.82
S5	Chernozem	Highway A 1	Rostovka village	17.6	2,290	26.8	0.36	43.1	196	67.9	2.34E3	107	41.8	189	7.45	3.06
S6	Chernozem	Highway A 343	Kokshetau town	35.6	135,106	21.0	0.26	26.1	133	44.9	1.18E3	74.4	26.6	130	7.37	2.81
S7	Chernozem	Highway A 1	Kokshetau town	1.3	135,106	29.4	0.46	44.6	217	83.4	2.18E3	112	40.0	169	5.76	2.16
S8	Chernozem	Highway A 1	Kenesary village	1.4	1,598	32.7	0.42	55.4	185	86.9	2.11E3	119	40.0	181	7.24	2.41
S9	Chernozem	Highway A 1	Shchuchinsk town	6.3	44,106	42.0	0.51	44.9	192	80.1	2.42E3	96.3	39.6	182	6.33	3.07
S10	Chernozem	Highway A 1, Gas station "Sinooil"	Makinka village	2.3	2,013	23.5	0.78	31.8	200	51.1	3.30E3	73.1	70.9	210	5.32	1.63
S11	Chernozem	Highway A 1	Akkol village	17.5	14,217	17.4	0.31	54.1	109	55.1	3.00E3	58.8	161	128	7.46	1.40
S12	Kastanozem	Highway A 1	Bazoygir village	3.3	2,526	26.6	0.29	34.6	131	53.4	2.07E3	87.5	42.9	140	7.53	2.10
S13	Kastanozem	Highway M 36	Astana city	0	613,006	16.5	0.25	18.4	79.5	38.5	1.09E3	53.4	23.8	109	7.34	1.96
S14	Kastanozem	Highway M 36	Anar village	29.4	1,117	27.7	0.34	26.5	128	54.2	1.43E3	69.1	30.3	135	7.69	2.19
S15	Kastanozem	Highway M 36	Anar village	1.4	1,117	24.1	0.40	22.0	112	43.4	1.31E3	60.7	31.8	115	6.51	1.62
S16	Kastanozem	Highway M 36	Temirtau town	22	169,590	8.9	0.21	15.3	72.6	38.8	982	45.5	31.5	134	7.44	2.36
S17	Kastanozem	Highway M 36	Karaganda town	3.5	459,778	13.5	0.31	16.4	87.6	37.4	1.15E3	49.2	27.0	132	7.26	2.00
S18	Calcisol + Solonetz	Highway M 36	Akbastau village	17.6	3,300	18.8	0.29	19.2	92.6	40.6	1.38E3	52.8	30.4	130	6.30	1.34
S19	Calcisol + Solonetz	Highway M 36	Aksu-Ayuly village	10.9	4,586	32.5	0.49	32.6	157	90.1	1.44E3	86.0	61.8	264	6.91	0.92
S20	Calcisol + Solonetz	Highway M 36	Aksu-Ayuly village	35.1	4,586	14.0	0.17	14.9	71.5	33.2	966	38.4	25.6	98.7	6.76	1.30
S21	Arenosol	Highway M 36	Akshatau village	10.4	1,149	13.8	0.26	16.7	80.4	40.4	1.02E3	47.6	20.3	109	6.62	0.98

S22	Arenosol	Highway M 36	Akzhal village	22.8	3,397	24.5	0.24	16.5	129	32.7	1.16E3	57.2	26.4	110	7.71	0.69
S23	Arenosol + Solonetz	Highway M 36	Balkhash town	55.6	68,883	14.9	0.12	16.3	68.6	36.1	1.07E3	48.4	23.6	92.3	7.99	0.59
S24	Arenosol + Solonetz	Highway M 36	Konirat village	3.1	3,103	13.1	0.73	14.4	116	25.6	353	33.3	8.6	23.6	7.91	0.62
S25	Arenosol + Solonetz	Highway M 36	Balkhash lake, recreation area Gulf Stream, Balkhash town	28.9	68,883	8.70	0.14	10.8	44.8	26.4	854	35.0	13.6	80.9	8.35	0.61
S26	Arenosol	Highway M 36	Balkhash lake, Balkhash town	64.4	68,883	14.4	0.15	17.6	69.4	37.3	1.09E3	44.7	32.1	107	8.40	0.52
S27	Solonchak + Solonetz	Highway M 36	Saryshagan village	15.7	4,429	17.8	0.45	23.4	100	52.6	1.71E3	59.7	39.3	135	8.14	0.67
S28	Arenosol	Highway M 36	Akbakay village	72.7	1,473	24.0	0.52	22.0	90.0	105	1.76E3	51.8	48.9	144	8.53	0.43
S29	Arenosol	Highway M 36	Balkhash lake chemical plant "Jambul Cement"	9.2	721	20.6	0.32	22.8	110	68.6	1.45E3	50.7	48.1	124	9.18	0.45
S30	Arenosol	Highway M 36	Shyganak village	13.9	2,402	16.8	0.40	18.3	83.3	42.7	1.18E3	53.3	36.7	136	8.65	0.38
S31	Arenosol	Highway M 36	Aksuek village	42.1	1,231	11.7	0.36	13.8	33.7	66.3	726	23.0	27.9	127	8.25	0.87
S32	Arenosol	Highway A 358	Birlik village	16.7	3,157	16.8	0.85	30.6	127	85.6	2.88E3	65.7	44.7	243	8.76	0.77
S33	Chemozem + Solonetz	Highway A 358	Kenes village	0.5	2,332	16.2	0.36	33.9	157	44.2	1.91E3	99.1	31.4	161	7.96	1.00
S34	Regosol	Highway A 358	Chu river 200m, Tole Bi village	1.3	19,000	20.4	0.29	24.5	97.7	43.7	1.69E3	63.4	32.9	117	7.35	2.07
S35	Regosol	Highway P 29	Aspara village	5.1	1,086	17.5	0.43	75.5	143	58.2	1.90E3	104	32.7	134	8.18	1.69
S36	Regosol	Highway P 29	Oytal village	13.4	4,181	33.0	0.76	80.6	190	91.4	3.84E3	159	65.3	195	7.78	3.61
S37	Umbrisol	Highway A 2	Zhambul village	4.8	6,633	29.0	0.53	36.4	163	63.5	2.13E3	97.7	46.9	191	8.33	2.48
S38	Umbrisol	Highway A 2	Kokdonen village	0.5	2,063	21.4	0.29	29.6	104	44.9	1.76E3	72.5	36.3	146	8.27	2.46
S39	Umbrisol	Highway A 2	Terenozek village	4.7	2,060	31.4	0.59	37.8	180	68.3	2.06E3	103	48.5	166	7.83	2.15
S40	Arenosol	Highway A 2, Gas station "Sinooil"	Talas village	2.3	2,921	28.0	0.51	39.8	144	60.2	2.71E3	87.8	51.0	181.3	7.70	2.80